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RECENT DEVELOPMENTS IN THE STUDY  
OF RADIOACTIVE SUBSTANCES.\*

BARELY eight years have elapsed since the discovery of Becquerel rays. Yet during that time the subject of radioactivity has developed so rapidly that it has now become an important branch of physics and chemistry. The phenomena are interesting in themselves, in some cases almost startling. But even more important is the bearing of the results upon some of the conceptions that lie at the very foundation of physical science. The study of radioactivity seems destined to exert a profound influence upon physical and chemical theories.

Without entering into the history of the subject, I shall first call attention to the results that are now best established, putting the facts into as systematic form as possible. The contradictory character of the early work, and the great complexity of the phenomena themselves, make this as difficult as it is desirable.

A radioactive substance may be defined as a substance which sends out Becquerel rays; *i. e.*, rays that are capable of penetrating bodies usually regarded as opaque, and which produce certain characteristic photographic and electric effects. In their general behavior such rays show a close resemblance to Roentgen rays; the differences will be referred to later. In the table below is given a list of the radioactive substances now known.

RADIOACTIVE SUBSTANCES.

*Permanently Active.*

Uranium .....	(238)
Thorium .....	(232)
Radium .....	(225†)

\* Address delivered before the Cornell Section of the American Chemical Society on May 18, 1903.

† A study of the spectrum of radium has led Runge and Precht to assign to this new element the atomic weight 25.8 instead of the value 22.5

Polonium..... (radioactive bismuth)  
Actinium.  
Radioactive lead.

*Temporarily Active.*

Ur-X, Th-X, excited activity obtained from air, from freshly fallen rain or snow, or from permanently active bodies.

It will be noticed that the list is divided into three groups. The first, containing uranium, thorium and radium, consists of elements whose separate existence is well established. This statement may now be made in the case of radium as well as in the case of the other two, since this substance has recently been so completely isolated as to make possible the determination of its atomic weight, while it has been shown by several observers to possess a characteristic spectrum. It will be noticed that the elements in this radioactive group possess atomic weights greater than any other known elements.

The second group is made up of suspected new elements. These elements have not been isolated and have not yet been found to give characteristic spectra. It is thought by some that the radioactivity in these cases is due to the presence of a trace of radium—so small as not to be detected by an ordinary test. It appears to me that the arguments against this view are strong. But the question can only be settled by more extended experimental study.

In the case of the substances of the first two groups, with the possible exception of polonium, the radioactivity is permanent so far as our present knowledge goes. In other words, these substances continue to give out Becquerel rays without special stimulation, such as is required for ordinary phosphorescence, and with no diminution in intensity that has thus far been detected. The question as to whether any obtained by Madame Curie by chemical methods. Some uncertainty therefore still exists.

substances are permanently active in a strict sense is not to be regarded as settled. I think, however, that most physicists feel that all active substances must gradually lose their activity, even though the rate of loss is too small to have been yet detected.

A third group of substances contains those which possess temporary activity, lasting for a period ranging from a few minutes to several months. Temporary activity may be acquired in a large number of ways. It may be obtained from the atmosphere, from freshly fallen rain or snow, from certain products developed by chemical processes from other active substances, and in a variety of other ways.

In dealing with the effects of the rays produced by radioactive substances we may conveniently adopt the classification shown in the list given below.

#### EFFECTS OF BECQUEREL RAYS.

*Photographic action.*

*Electric effects.* (The most important of these is the power possessed by the rays of making air and other gases temporarily conducting.)

*Luminous effects.* (Fluorescence produced by the rays in various substances.)

*Chemical effects.* (Development of ozone, color changes produced in glass, etc.)

*Physiological effects.* (Burns produced by long exposure to the rays; sensation of light produced by highly active preparations held near the eye.)

Like Roentgen rays, the rays sent out by radioactive substances do not show regular reflection or refraction.

It will be noticed that practically all of the effects produced by Becquerel rays are also produced by X-rays. It would be natural to conclude that the rays are of the same type. Yet there are enough differences in the properties of the two rays to show that this can not be true. For example, Becquerel rays are deflected by a magnetic field and by an electric field and carry an electric charge. X-rays possess

none of these properties. It is probable that a radioactive substance sends out *some* X-rays; but the bulk of the rays emitted by it are of a different kind.

The methods used in studying Becquerel rays are naturally based upon the various effects which these rays produce. Up to the present time the photographic effect and the electrical effect have been the ones chiefly employed in the study of the rays. Of the two the electrical method is capable of far greater sensitiveness. In brief, this method is applied in the following way: An insulated conductor of small capacity is connected with a sensitive electrometer and is then charged to a potential of one to two hundred volts. This conductor is placed in a metallic vessel whose walls are grounded. With good insulation the conductor will hold its charge under ordinary circumstances for a long period; but if Becquerel rays are allowed to enter the vessel they make the air a conductor and thus permit the charge to escape. The rate at which the charge escapes, as indicated by the electrometer, is a measure of the intensity of the rays.

In the early study of the subject different observers often obtained contradictory results. In many cases the contradictions have since been explained by the fact that some used the photographic method while others used the electrical method. The two methods of measuring the intensity of the rays do not agree. For example, a substance *A* may produce very strong photographic effects, while another substance *B*, also tested photographically, is found to give out rays that are relatively weak. But if the two substances are compared by means of the electrical effects which they produce, it may turn out that *B* is more active than *A*.

Results such as this have led to the conclusion that there are different kinds of

Becquerel rays, some of which produce strong photographic effects, while others, not so strong photographically, produce intense electrical effects. More recently it has been found not only that the rays from different substances differ, but that a single substance sends out rays of widely different properties. Three types of Becquerel rays have thus far been recognized. An active substance in general sends out all three kinds, but the distribution of the radiation among the different types depends on the substance. For convenience these rays have been referred to as the  $\alpha$ ,  $\beta$  and  $\gamma$  rays. A brief statement of more important properties of each kind of ray is given in the accompanying table.

#### DIFFERENT TYPES OF RAYS.

##### *$\alpha$ Rays.*

Readily absorbed (*e. g.*, by a thin sheet of aluminium foil, or even by a few centimeters of air).

Relatively strong electrically, *i. e.*, in making gases conducting.

Photographic effect small.

Behavior in an electric field and in magnetic field such as to indicate that these rays are positively charged particles of molecular dimensions moving at a speed of about  $10^9$  cm./sec.

##### *$\beta$ Rays.*

Quite penetrating (*e. g.*, pass through several millimeters of aluminium or glass).

Electrical effects weak.

Photographic effects relatively strong.

Carry a negative charge.

Probably consist of negatively charged particles, much smaller than atoms, moving at a speed nearly equal to the speed of light, *i. e.*,  $3.10^{10}$  cm./sec. Behavior in electric and magnetic field consistent with this view.

##### *$\gamma$ Rays.*

Highly penetrating. Pass through several centimeters of metal.

Probably Roentgen rays.

The existence of these different kinds of rays may be proved and their separation may be effected in a number of different

ways. The most obvious way is by means of absorption. For example, a layer of aluminium foil will absorb practically all of the  $\alpha$  rays, while it permits the  $\beta$  rays to pass with scarcely any diminution in intensity. To separate the  $\beta$  rays from the  $\gamma$  rays by absorption is more difficult, for both of these rays are highly penetrating. Separation may here be effected, however, by passing the rays through a magnetic field, since the  $\beta$  rays are deflected, while the  $\gamma$  rays are not.

Let us consider first the  $\beta$  rays. Practically all physicists now agree in regarding these rays as consisting of very small negatively charged particles, or electrons, moving at great speed. That they carry a negative charge has been shown by direct experiment. It is also an experimental fact that these rays are deviated in a magnetic field, and in an electric field in the manner that would be expected if they were charged particles in motion. But the quantitative relations are such as to indicate that if this hypothesis is correct the mass of each particle must be much less than the mass of the smallest atom, while the speed of the particles must be nearly the speed of light. Each of these statements seems so incredible and revolutionary that it is difficult to accept the hypothesis, even in spite of its complete agreement with experiment at every point where a test can be applied. I think that the difficulty in accepting this hypothesis is probably greater in the minds of chemists than it is with physicists. The battle over the electron theory in its purely physical aspects had already been fought out during the development of the theory of cathode rays.\* The behavior of the  $\beta$  rays is so similar to that of cathode rays,

\* The development of this subject has been traced by the writer in an article on 'Cathode Rays and some Related Phenomena,' SCIENCE, Vol. XII., p. 41, 1900.

that the electron theory, which has now been universally accepted in the case of the cathode rays, has naturally been extended to include the Becquerel rays.

Until recently the  $\alpha$  rays were supposed to be undeflected in passing through a magnetic field. Within the last few months, however, it has been shown both by Rutherford and Becquerel that these rays do experience a deflection. This deflection is in the opposite direction from that experienced by the  $\beta$  rays, and is much less in amount. An extremely strong field is necessary to show the deflection at all. This behavior of  $\alpha$  rays is explained by assuming the rays to consist of positively charged particles moving at high speeds. It appears, however, that the particles are much larger, that the speed of these rays, instead of being nearly that of light, is only about one tenth as great.

The third type of rays, the  $\gamma$  rays, has been only slightly studied; but so far as investigation has proceeded the properties of the  $\gamma$  rays are the same as those of X-rays. These rays are highly penetrating and have been found to pass through several centimeters of metal.

The intensity of the rays from radium is much greater than that of the rays from the other active substances. Nearly pure radium preparations have been made which show an activity, as measured by the electrical effect, five hundred thousand times as great as the activity of metallic uranium. Small traces of radium in minerals thus add greatly to the activity of these minerals, even when the amount present is so small that no chemical test can detect it. The electrical and photographic methods of testing for radioactivity are, in fact, more sensitive than any chemical or spectroscopic tests yet discovered.

The great difference between the activity of radium and that of the other active sub-

stances, and the fact that small traces of radium are hard to detect, has led to the thought that the activity of other substances might be due to the presence as an impurity of some highly active element, possibly radium itself. In the case of uranium it seemed for a time as though this view was definitely confirmed. Upon precipitating a solution of uranium by ammonium carbonate Crookes succeeded in separating uranium into two parts, one of which was redissolved by excess of the reagent, while the other remained behind as a precipitate. Only a trace of the latter was obtained, but it was many times more active than the original uranium. In fact, when tested by photographic methods this uranium-X, as it is called, seemed to have *all* of the original activity, while the ordinary uranium was no longer active. It was found, however, that the uranium-X soon lost its activity, falling to one half its original strength in about twenty-two days; while the ordinary uranium gradually recovered its activity, regaining one half of its original strength in about the same length of time.

The investigation which led to the separation of uranium-X illustrates the contradictions which may arise in work of this kind. Crookes used the photographic method and obtained the results just stated. Others, repeating his work by the electrical method, reached very different conclusions. When tested electrically the increased activity of Ur-X was not nearly so marked, while the ordinary uranium was nearly as active as ever. The separation is now seen to be one which divides the active uranium into two parts, both of which are active; but one part gives out  $\alpha$  rays, while the other gives chiefly  $\beta$  rays. A similar separation has been effected in the case of thorium. In this case thorium nitrate is precipitated

from solution by ammonium hydrate. The filtrate, which is free from thorium, when evaporated to dryness shows an activity measured electrically fully one thousand times as great as that of the original thorium. Thorium-X, like Ur-X, is present as a trace only. Its radiation consists chiefly of  $\beta$  rays. But both thorium and Th-X develop rays of both kinds.

It was early found that both thorium and radium possess the power of exciting temporary activity in bodies placed near them. This excited activity may also be produced by bringing air that has been in contact with radium or thorium past the body to be excited. The results are explained by the fact that thorium and radium each give out an 'emanation,' which behaves in all respects like an inert gas. This emanation is itself radioactive, as may be shown by the electrical effects produced by it, and it also has the power of exciting temporary activity in bodies with which it comes in contact. The emanations lose their activity rather rapidly; in the case of thorium the activity falls to one half its original value in about one minute, while in the case of radium a similar loss occurs in the course of several days. Radium and thorium are the only substances that give emanations; they are also the only substances which have the power of exciting activity in neighboring bodies.

That the emanations of radium and thorium are gases is confirmed in a great variety of ways. For example, they can be occluded by porous solids, such as the solid salts which develop them. Owing to the fact that the radium emanation preserves its activity for a long time, the occlusion of this emanation is more readily studied. A large part of emanation developed by radium is occluded by the radium salt itself; this may be driven off

by heating, after which considerable time is required for the original condition to be restored. When the active salts of radium or thorium are in solution the emanations developed are liberated more rapidly, there being in this case no chance for occlusion. The rate at which the emanation is developed appears to be constant under all conditions.

The emanations of both radium and thorium are chemically inert. They may be passed through sulphuric acid, nitric acid and hydrochloric acid without change, and are also unaffected by passing over red-hot lead chromate or magnesium. They may be condensed, however, by passing through a tube immersed in liquid air. The radium emanation condenses at  $-150^{\circ}$  C., and that of thorium at about  $-120^{\circ}$  C. The rate of decay of the emanation is unaffected by this low temperature. When the temperature is raised again the emanations are liberated with an activity depending only upon the time that has elapsed since they were developed.

The excited activity produced by the emanations of radium and thorium is greatest on bodies that are negatively charged. It would seem, therefore, that the substance to which excited activity is due must itself be positively charged. If the substance is sending out more  $\beta$  rays than  $\alpha$  rays such a positive charge would naturally follow.

Temporary activity may be acquired by exposing a negatively charged body to ordinary air. Apparently the atmosphere contains a radioactive gas similar to the emanations mentioned above. This view is strengthened by the fact that freshly fallen rain and snow possess temporary activity, probably obtained from the air. Air from cellars and air that has been drawn from a porous soil are especially rich in this active constituent. J. J.

Thomson has recently found that the water from certain deep wells in Cambridge also contains a radioactive gas.

Several general points brought out in recent years should not be lost sight of, since their bearing upon the theory of the subject seems to be important.

Radioactivity seems to be unaffected by temperature. Neither the activity of a substance nor the rate of decay in the case of temporary activity is affected by a change of temperature as great as that from liquid air to a white heat. None of the physical agencies which usually affect physical phenomena seems to influence radioactivity. These facts, together with the fact that the salts of the active elements are active as well as the elements themselves, have suggested the thought that the phenomena of radioactivity are phenomena of the atom rather than of the molecule.

There are many indications that radioactivity is accompanied by some change in the active substances. For example, the Th-X may be removed from thorium completely; but at the end of a month or so the thorium may be again treated in the same way and as much Th-X obtained as before. It seems as though the development of Th-X was proceeding slowly all the time. The Th-X must itself be undergoing some change, since its activity diminishes steadily from the time of its separation. Further evidences of change are furnished by the continual development of the emanations of thorium and radium and the gradual decay of the activity possessed by these emanations.

Two questions of the greatest importance at once suggest themselves in the consideration of radioactive substances. First of all, if the rays consist of particles shot off from the substances it would seem as though a diminution in weight should re-

sult. It seems hardly probable that matter could be gathered in from the surroundings to supply this loss. There is in fact some experimental evidence that strong active preparations lose in weight by a measurable amount; but this evidence is not yet to be regarded as conclusive. Observations to test this point are difficult. The change to be expected is extremely minute, even with the most active substances, and might easily be masked by the results of chemical changes produced by the rays in the walls of the containing vessel. But although the question must be regarded as unsettled, I think that most students of the subject are convinced that a loss of weight actually occurs, even though it has not as yet been detected.

A question of even more fundamental importance, and one that is now attracting especial interest, is that of the source of the energy which the rays possess. It has recently been shown by the Curies that one gram of an active radium preparation develops each hour forty calories of energy. In other words, a gram of this preparation could melt its own weight of ice in two hours. Expressed in a different form, this would mean that this salt of radium develops, in the course of a month, as much energy as is liberated by the combustion of an equal mass of hydrogen. When hydrogen is burned its store of energy is exhausted; but radium can apparently continue to give out this energy month after month, with no diminution in intensity which has yet been detected. The numerical results obtained by the Curies may require correction in the light of more accurate measurements; but the difficulty will still remain. How is it possible for a radioactive substance to continue radiating energy for an indefinite period without appreciable loss?

Several explanations have been suggested.

It has been thought, for example, that radioactive substances may have the power of absorbing the energy of certain rays, hitherto undetected, which are all the while proceeding through space, coming perhaps from the sun. The radioactive substances might utilize the energy thus absorbed in the development of Becquerel rays, just as fluorescent substances utilize the energy absorbed from sunlight in producing luminescent phenomena. This view, I believe, is supported by the Curies. Sir Wm. Crookes has suggested that radioactive substances possess the power of utilizing the energy of surrounding bodies. Such an explanation might possibly contradict the second law of thermodynamics; yet, since the process is not a cyclic one, I do not believe that any contradiction would be found. An objection to this explanation has been raised, based on the fact that radioactivity continues when the substance is placed in a vacuum. Crookes replies, however, that the best vacuum ever obtained contains millions of molecules per cubic centimeter, so that enough are left to supply the energy needed. A test might be applied, as suggested by J. J. Thomson, by placing the active substance in an ice calorimeter entirely surrounded by ice. If the ice melts, as is probable, it would seem that the Crookes explanation could not hold.

Elster and Geitel suggested, a few years ago, that the energy might be derived from processes of molecular or atomic change which accompany the development of Becquerel rays. According to this view, an active substance is one which is slowly changing from an unstable condition into a more permanent form; the processes which go on during this change may bring about the development of Becquerel rays, while the energy developed is that liberated during the transition. The produc-

tion of Th-X from Th is perhaps the first stage in such a change; the decay in the activity of the Th-X and the production of the emanation, perhaps form the second stage; while a third step in the progressive alteration of the original substance is shown by the development of excited activity from the emanation. The final products of this disintegration process are doubtless much simpler in their structure than the original substance, and are probably not radioactive. It has been suggested that helium, which has so far been obtained only from radioactive minerals, may be one of the final products of radioactive change.

Perhaps the most serious difficulty in accepting the explanation just mentioned arises from the large quantities of energy involved. It seems almost incredible that so much energy could be stored in a few milligrams of material. The changes assumed are, however, atomic changes. May it not be that such changes involve energy quantities of a higher order? As we proceed from the ordinary motions of mass mechanics toward motions of a finer grain the energy involved increases. A falling raindrop possesses energy due to its mass motion; the energy liberated when it freezes is much greater; and the energy involved in the formation of the drop from oxygen and hydrogen is greater still. May not the energy of atomic synthesis and disintegration be as much greater than that of ordinary chemical change as the latter is greater than the energy of physical change? The view advanced by Elster and Geitel appears to me to give the best explanation that has yet been offered. But this question of the energy of the rays, like many other questions that have been raised by the study of radioactivity, can by no means be looked upon as settled.

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